



(11) Publication number : **0 683 220 A2**

(12)

EUROPEAN PATENT APPLICATION

(21) Application number : **95303343.8**

(51) Int. Cl.⁸ : **C10M 159/12, C10M 139/00,
C10M 177/00, // C10N40:04**

(22) Date of filing : **18.05.95**

(30) Priority : **18.05.94 US 245600**

(43) Date of publication of application :
22.11.95 Bulletin 95/47

(84) Designated Contracting States :
BE DE ES FR GB IT

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(54) **Lubricant additive compositions.**

(57) Multifunctional additives having a variety of advantageous properties are described. They are formed by a process which comprises heating concurrently or in any sequence a) an oil-soluble ashless dispersant containing basic nitrogen with b) an alkoxyated alcohol and c) a borating agent in proportions on an active ingredient basis of about 0.03 to about 0.35 part by weight of b) per part by weight of a), and about 0.005 to about 0.06 part by weight of boron as c) per part by weight of a) to a temperature in the range of about 50 to about 150°C, and if water and/or solids are present in the resultant mixture, removing both of them or whichever of them is present in the resultant mixture. Preferably, another component is used in the formation of these additives, this component being an inorganic oxy acid or anhydride phosphorus used in proportions on an active ingredient basis of about 0.0005 to about 0.03 part by weight of phosphorus as the component per part by weight of a).

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TECHNICAL FIELD

This invention relates to novel metal-free boron-containing compositions that are useful as multifunctional additives in oils of lubricating viscosity. In addition, this invention relates to novel metal-free boron- and phosphorus-containing compositions that are useful as multifunctional additives in oils of lubricating viscosity.

BACKGROUND

Heretofore the formation of borated ashless dispersants has been reported. Typical disclosures on this subject include U.S.

3,087,936; 3,254,025; 3,281,428; 3,282,955; 3,284,409; 3,284,410; 3,338,832; 3,344,069; 3,533,945; 3,658,836; 3,703,536; 3,718,663; 4,455,243; and 4,652,387.

U.S. 4,857,214 describes oil-soluble lubricant additive compositions formed by heating an inorganic phosphorus acid or anhydride, including partial and total sulfur analogs thereof, a boron compound, and an ashless dispersant which contains basic nitrogen and/or a hydroxyl group. Such additives have excellent antiwear/EP properties and impart antioxidancy to lubricating oils including functional fluids such as automatic transmission fluids. In addition, such additives exhibit improved rubber seal protective properties.

THE INVENTION

Novel and eminently useful multifunctional additives are provided by this invention. One embodiment of this invention provides multifunctional additive compositions formed by a process which comprises heating concurrently or in any sequence a) an oil-soluble ashless dispersant containing basic nitrogen with b) an alkoxyated alcohol and c) a borating agent to a temperature in the range of about 50 to about 150°C, and if water and/or solids are present in the resultant mixture, removing both of them or whichever of them is present in the resultant mixture. These novel multifunctional additives once prepared and filtered are clear mixtures that tend to remain free of haze and solids even after long periods of storage at elevated temperatures such as 70°C, even though they may contain high levels of boron within the range of proportions described hereinafter.

Another embodiment provides multifunctional additives which are formed by heating concurrently or in any sequence a) an oil-soluble ashless dispersant containing basic nitrogen with b) an alkoxyated alcohol, c) a borating agent and d) an inorganic oxyacid or anhydride of phosphorus to a temperature in the range of about 50 to about 150°C, and if water and/or solids are present in the resultant mixture, removing

both of them or whichever of them is present in the resultant mixture. These novel additives possess the above storage stability properties as well as all of the advantageous properties of the additives described in U.S. 4,857,214. Indeed these additives can contain a high loading of both boron and phosphorus within the proportion ranges set forth hereinafter and remain stable for long periods of time at elevated temperatures without incompatibility problems being encountered. In addition, they exhibit improved performance in the Mitsubishi Water Test or the ASTM D 2711 demulsibility test as compared to a corresponding additive made in the same way with the same amount of the same materials except for the elimination of component b).

Lubricating oil compositions which comprise from 0.1 to 99.9 parts by weight of any of the above additive compositions and from 99.9 to 0.1 parts by weight of at least one oil of lubricating viscosity are additional embodiments of this invention. Another embodiment are lubricating oil compositions which comprise from 0.1 to 50 parts by weight of any of the above additive compositions and from 50 to 0.1 parts by weight of at least one oil of lubricating viscosity.

Other embodiments and features of this invention will become still further apparent from the ensuing description and appended claims.

Component a)

Basic nitrogen-containing ashless dispersants to which this invention is applicable include hydrocarbyl succinimides; hydrocarbyl succinamides; mixed ester/amides of hydrocarbyl-substituted succinic acids formed by reacting a hydrocarbyl-substituted succinic acylating agent stepwise or with a mixture of alcohols and amines, and/or with aminoalcohols; Mannich condensation products of hydrocarbyl-substituted phenols, formaldehyde and polyamines; and amine dispersants formed by reacting high molecular weight aliphatic or alicyclic halides with amines, such as polyalkylene polyamines. Mixtures of such dispersants can also be used.

Such basic nitrogen-containing ashless dispersants are well known lubricating oil additives, and methods for their preparation are extensively described in the patent literature. For example, hydrocarbyl-substituted succinimides and succinamides and methods for their preparation are described, for example, in U.S. Pat. Nos. 3,018,247; 3,018,250; 3,018,291; 3,172,892; 3,185,704; 3,219,666; 3,272,746; 3,361,673; and 4,234,435. Mixed ester-amides of hydrocarbyl-substituted succinic acid are described, for example, in U.S. Pat. Nos. 3,576,743; 4,234,435 and 4,873,009. Mannich dispersants, which are condensation products of hydrocarbyl-substituted phenols, formaldehyde and polyamines are described, for example, in U.S. Pat. Nos. 3,368,972;

3,413,347; 3,539,633; 3,697,574; 3,725,277; 3,725,480; 3,726,882; 3,798,247; 3,803,039; 3,985,802; 4,231,759 and 4,142,980. Amine dispersants and methods for their production from high molecular weight aliphatic or alicyclic halides and amines are described, for example, in U.S. Pat. Nos. 3,275,554; 3,438,757; 3,454,555; and 3,565,804.

The ashless dispersant treated in accordance with this invention is preferably a hydrocarbyl succinimide, a hydrocarbyl succinic ester-amide or a Mannich base of a polyamine, formaldehyde and a hydrocarbyl phenol in which the hydrocarbyl substituent is a hydrogenated or unhydrogenated polyolefin group and preferably a polypropylene or polyisobutene group having a number average molecular weight (as measured by gel permeation chromatography) of from 250 to 10,000, and more preferably from 500 to 5,000, and most preferably from 750 to 2,500. The ashless dispersant is most preferably an alkenyl succinimide such as is available commercially from Ethyl Petroleum Additives, Inc. and Ethyl Petroleum Additives, Ltd. as HITEC® 644 and HITEC® 646 additives and especially as HITEC® 634 additive. Other suitable commercially available alkenyl succinimides which may be used in the practice of this invention include for example Anglamol 890, 894, 935, 6406, 6418 and 6420 from The Lubrizol Corporation, ECA 4360, 5017, and 5025 from Exxon Chemicals, and OLOA 373, 374 and 1200 from Chevron Chemical Company.

In general amines containing basic nitrogen or basic nitrogen and additionally one or more hydroxyl groups, including amines of the types described in U.S. 4,235,435 can be used in the formation of the ashless dispersants. Usually, the amines are polyamines such as polyalkylene polyamines, hydroxy-substituted polyamines and polyoxyalkylene polyamines. Examples of polyalkylene polyamines include diethylene triamine, triethylene tetramine, tetraethylene pentamine, pentaethylene hexamine, and dipropylene triamine. While pure polyethylene polyamines can be used, it is generally preferred to use mixtures of linear, branched and cyclic polyethylene polyamines having an average in the range of about 2.5 to about 7.5 nitrogen atoms per molecule and more preferably an average in the range of about 3 to about 5 nitrogen atoms per molecule. Mixtures of this type are available as articles of commerce. Hydroxy-substituted amines include N-hydroxyalkyl-alkylene polyamines such as N-(2-hydroxyethyl)ethylene diamine, N-(2-hydroxyethyl)piperazine, and N-hydroxyalkylated alkylene diamines of the type described in U.S. 4,873,009. Polyoxyalkylene polyamines typically include polyoxyethylene and polyoxypropylene diamines and triamines having average molecular weights in the range of 200 to 2500. Products of this type are available under the Jeffamine trademark.

Component b)

Alkoxyated alcohols which can be used in forming the additives of this invention include oil-soluble alkoxyated alkanols, alkoxyated cycloalkanols, alkoxyated polyols, alkoxyated phenols, and alkoxyated heterocyclic alcohols which contain an average of up to about 20 alkoxy groups per molecule. The alkoxy groups can be ethoxy, propoxy, butoxy, or pentoxy, or combinations of two or more of these. However ethoxy-substituted alcohols are preferred. For best results, the alkoxyated alcohol should be a liquid at ambient temperatures in the range of 20-25°C. Since the alkoxyated alcohol should be oil-soluble, short chain alcohols preferably contains an average of at least two alkoxy groups per molecule whereas longer chain alcohols may contain one or more alkoxy groups per molecule. The average number of alkoxy groups in any given alcohol can be as high as 15 or 20 as long as the product is oil soluble and is preferably a liquid at room temperature. Examples of alcohols that form suitable alkoxyated alcohols include C₁₋₂₄ alkanols, C₁₋₁₀ cycloalkanols, polyols having up to about 16 carbon atoms and 2-5 hydroxyl groups, polyol ethers having up to about 16 carbon atoms and at least one hydroxyl group, phenol, alkylphenols having up to about 16 carbon atoms, and hydroxy-substituted heterocyclic compounds such as tetrahydrofurfuryl alcohol and tetrahydropyran-2-methanol.

Preferred is an ethoxyated C₈-C₁₆ alcohol or mixture of two or more of such alcohols having an average in the range of 1 to 10 ethoxy groups per molecule. Particularly preferred is an ethoxyated C₁₂ alcohol having average in the range of 1 to 3 ethoxy groups per molecule.

Component c)

Suitable boron materials which can be used as borating agents include boron acids, boron oxides, boron esters, ammonium borate, and super-borated dispersants -- i.e.; dispersants such as described above which have been heavily borated. Aminoboranes can be used, but are expensive. Boron halides, while useable, tend to contribute a halogen content to the dispersant or increase the halogen content of the dispersant, neither of which is desirable.

Examples of suitable borating agents include boron acids such as boric acid, boronic acid, tetraboric acid, metaboric acid, pyroboric acid, esters of such acids, such as mono-, di- and tri-organic esters with alcohols having 1 to 20 carbon atoms and/or phenols having 6 to 20 carbon atoms, e.g., methanol, ethanol, isopropyl alcohol, butanols, pentanols, hexanols, cyclopentanol, cyclohexanol, methylcyclohexanol, ethylene glycol, propylene glycol, phenol, cresols, xylenols, and the like, and boron oxides such as boron ox-

ide and boron oxide hydrate. Orthoboric acid is a preferred borating agent for use in the practice of this invention.

Component d)

Examples of inorganic phosphorus acids and anhydrides which are useful in forming the preferred products of the invention include phosphorous acid (H_3PO_3), pyrophosphorous acid, phosphoric acid, hypophosphoric acid, phosphorus trioxide (P_2O_3), phosphorus tetraoxide (P_2O_4), and phosphoric anhydride (P_2O_5), also known as phosphorus pentoxide). Mixtures of two or more such compounds can be used. Preferred is phosphorous acid (H_3PO_3).

Proportions

Components a), b) and c) are typically used in proportions on an active ingredient basis of about 0.03 to about 0.35 part by weight of b) per part by weight of a), and about 0.005 to about 0.06 part by weight of boron as c) per part by weight of a). Preferably, these proportions on the same basis are from about 0.08 to about 0.30 part by weight of b) and about 0.01 to about 0.05 part by weight of boron as c) per part by weight of a). More preferred proportions are from about 0.12 to about 0.25 part by weight of b) and about 0.012 to about 0.025 part by weight of boron as c) per part by weight of a). Especially preferred proportions are from about 0.15 to about 0.20 part by weight of b) and about 0.015 to about 0.02 part by weight of boron as c) per part by weight of a). When component d) is included in the mixture being heated the proportions are such that on an active ingredient basis there is by weight from about 0.0005 to about 0.03 part, preferably from about 0.001 to about 0.025 part, more preferably from about 0.005 to about 0.02 part, and most preferably from about 0.01 to about 0.02 part, of phosphorus as d) per part by weight of component a). By "on an active ingredient basis" is meant that the weight of any solvent or diluent used with any component is excluded from the calculation of proportions.

Reaction Conditions

As noted above, in one of its forms, this invention involves heating a basic nitrogen-containing ashless dispersant with components b) and c) above, or with components b), c) and d) above to produce a clear homogeneous additive composition. After charging component a), the other components being used can be heated separately with the reactor contents and in so doing, the other components -- i.e., components b) and c) or components b), c) and d) -- can be charged to the reaction vessel and heated therein in any sequence. When using components b), c) and d), these

components may be charged and heated in one or more sub-combinations in any sequence (e.g., b) + c), then d); b) + c) then b) + d); etc.). Preferably the components being used -- b) and c) or b), c) and d) -- are heated concurrently with component a).

The reactor contents are heated, preferably with agitation, at a suitably elevated temperature within the range of 50 to 150°C, preferably about 90 to 110°C, most preferably at about 100°C. The over-all time may vary from about 1 hour or less to about 6 hours or more depending on the temperature. If water is evolved, such as when using boric acid as component c), the water should be removed by distillation at reduced pressure. Any solids that may remain in the resultant mixture are then removed, preferably by filtration to produce a clear, oil-soluble product.

The process can be carried out in the absence of solvent by mixing and heating the reactants. Preferably, however, if an inorganic borating agent such as boric acid is used as component c), water is added to facilitate the initial dissolution of the boron compound. Water formed in the reaction and any added water is then removed by vacuum distillation at temperatures of from 100-140°C. Preferably, the reaction is carried out in a diluent oil or a solvent such as a mixture of aromatic hydrocarbons.

Optionally, additional sources of basic nitrogen can be included in the ashless dispersant mixture being used in the process so as to provide a molar amount (atomic proportion) of basic nitrogen up to that equal to the molar amount of basic nitrogen contributed by the ashless dispersant. Preferred auxiliary nitrogen compounds are long chain primary, secondary and tertiary alkyl amines containing from 12 to 24 carbon atoms, including their hydroxyalkyl and aminoalkyl derivatives. The long chain alkyl group may optionally contain one or more ether groups. Examples of suitable compounds are oleyl amine, N-oleyltrimethylenediamine, N-tallow diethanolamine, N,N-dimethyloleylamine and myristyl-oxypropyl amine.

Other materials normally used in lubricant additives which do not interfere with the reaction may also be added, for example, a benzotriazole, including lower (C_1 - C_4) alkyl-substituted benzotriazoles, which function to protect copper surfaces.

The amount of boron compound(s) preferably employed ranges from 0.001 mole to 1 mole per mole of basic nitrogen in the mixture up to one half of which may be contributed by an auxiliary nitrogen compound. The amount of phosphorus compound(s), when employed, ranges from 0.001 mole to 0.999 mole per mole of basic nitrogen in the mixture which is in excess of the molar amount of boron compound(s) used.

The amount of added water, when used, is not particularly critical as it is removed by distillation when no longer needed for improving solubility of an

inorganic borating agent. Amounts up to about one percent by weight of the mixture are preferred. When used, the amount of diluent generally ranges from 10 to 50 percent by weight of the mixture. When added, the amount of copper protectant generally ranges from 0.5 to 5 percent by weight of the mixture.

The following examples in which parts are by weight are illustrative.

EXAMPLE 1

A mixture of 86.61 parts of HITEC® 2605 additive (a Mannich base dispersant; Ethyl Corporation), 10.07 parts of ethoxylated lauryl alcohol (Trycol 5966; Henkel Corporation), 2.61 parts of 70% phosphorous acid, 5.90 parts of boric acid and 0.81 part of tolyltriazole (Cobratec TT-100) is heated with stirring at 110°C for 2 hours. Then the temperature is increased to 140°C and a vacuum of 40 mm is applied and the mixture is kept under these conditions for a period of one hour to remove water formed in the process. The product is then filtered. The resultant solids-free liquid additive contains about 0.41% phosphorus and about 1.03% of boron.

EXAMPLE 2

The procedure of Example 1 is repeated except that an equal weight of HITEC® 7100 additive (a succinic ester-amide dispersant; Ethyl Corporation) is used instead of the Mannich base dispersant.

The resultant additive composition has a phosphorus content of about 0.43% and a boron content of about 0.88%.

EXAMPLE 3

The procedure of Example 1 is repeated except that an equal weight of HITEC® 646 additive (a tetraethylene pentamine succinimide dispersant having a polyisobutenyl substituent formed from polyisobutene having a GPC number average molecular weight of about 1300; Ethyl Corporation) is used instead of the Mannich base dispersant. The resultant additive composition has a phosphorus content of about 0.44% and a boron content of about 1.04%.

EXAMPLES 4-6

The procedures of Examples 1-3 are repeated except that the phosphorous acid is omitted. The resultant additive compositions contain similar amounts of boron.

EXAMPLE 7

The procedure of Example 3 is repeated except that the succinimide used is a mixture of 30% of min-

eral oil and 70% of HITEC 634 additive (a succinimide dispersant formed from a polyisobutenyl succinic anhydride and triethylene tetramine in a mole ratio of about 1.8 moles of the anhydride per mole of triethylene tetramine, and wherein the polyisobutenyl group of the ashless dispersant is derived from polyisobutene having a GPC number average molecular weight in the range of about 850 to about 1000; Ethyl Corporation).

EXAMPLE 8

The procedure of Examples 7 is repeated except that the phosphorous acid is omitted.

EXAMPLE 9

The procedure of Example 7 is repeated using 83.3 parts of the 70% oil solution of the succinimide dispersant, 9.9 parts of the Trycol 5966, 5.8 parts of boric acid, 1.8 parts of phosphorous acid, 0.8 part of TT-100 and 0.7 part of water. Approximately 2.5 parts of water are recovered from the reduced pressure distillation.

EXAMPLES 10-11

Example 9 is repeated substituting in one case Sterox.ND (an ethoxylated alkyl phenol; Monsanto Corporation) and in another case Trycol 5940 for the Trycol 5966. Similar storage stable products are obtained.

EXAMPLES 12-14

Example 9 is again repeated using in one case twice the amount of phosphorous acid, in another case without use of the TT-100 and in another case using twice the amount of Trycol 5966. In each case, storage stable additive compositions are formed.

EXAMPLE 15

Example 9 is repeated except that the dispersant is HITEC® 645 additive (a succinimide dispersant formed from a polyisobutenylsuccinic anhydride and triethylene tetraline in a mole ratio of about 1.6 moles of the anhydride per mole of triethylene tetramine, and wherein the polyisobutylene group of the ashless dispersant is derived from polyisobutene having a GPC number average molecular weight in the range of about 850 to about 1000; Ethyl Corporation).

Additives made as in Examples 1-3 are not only effective as antiwear/extreme pressure additives but additionally contribute rust inhibition properties to formulated gear oils and gear oil additive packages. Additives made as in Examples 7 and 9-15 have good demulsibility properties and water compatibility as

shown for example by the Mitsubishi Water Test or the ASTM D 2711 demulsibility test. Additives made such as in Example 7 also improve the copper corrosion resistance of formulated gear oils. Moreover, the products of this invention have good thermal and oxidative stability and thus can be stored for long periods of time without haze or sediment formation, even though they contain high proportions of boron alone or high proportions of both boron and phosphorus. Another very desirable characteristic of the additives of this invention is that they do not contain any metal.

The additives of this invention can be utilized in combination with conventional gear oil additive components such as sulfur-containing antiwear agents, phosphorus-containing extreme pressure agents, copper corrosion inhibitors, rust inhibitors, foam inhibitors, oxidation inhibitors, demulsifiers, and the like. These can be used in their conventional concentrations in the finished gear oils.

As used herein the term "oil-soluble" means that the substance under discussion should be sufficiently soluble at 20°C in the base oil selected for use to reach at least the minimum concentration required to enable the substance to serve its intended function. Preferably the substance will have a substantially greater solubility in the base oil than this. However, the substance need not dissolve in the base oil in all proportions.

Each and every U.S. patent document referred to hereinabove is fully incorporated herein by reference.

Claims

1. An additive composition obtainable by a process which comprises heating concurrently or in any sequence
 - a) an oil-soluble ashless dispersant containing basic nitrogen with
 - b) an alkoxyated alcohol and
 - c) a borating agent in proportions on an active ingredient basis of 0.03 to 0.35 part by weight of b) per part by weight of a), and 0.005 to 0.06 part by weight of boron as c) per part by weight of a) to a temperature in the range of 50 to 150°C, and if water and/or solids are present in the resultant mixture, removing both of them or whichever of them is present in the resultant mixture.
2. An additive composition obtainable by a process which comprises heating concurrently or in any sequence
 - a) an oil-soluble ashless dispersant containing basic nitrogen with
 - b) an alkoxyated alcohol,
 - c) a borating agent and d) an inorganic oxyacid or anhydride of phosphorus in proportions

on an active ingredient basis of 0.03 to 0.35 part by weight of b) per part by weight of a), 0.005 to 0.06 part by weight of boron as c) per part by weight of a), and 0.0005 to 0.03 part by weight of phosphorus as d) per part by weight of a), to a temperature in the range of 50 to 150°C, and if water and/or solids are present in the resultant mixture, removing both of them or whichever of them is present in the resultant mixture.

3. A composition according to claim 2 wherein said proportions on an active ingredient basis are 0.12 to 0.25 part by weight of b) per part by weight of a), 0.012 to 0.025 part by weight of boron as c) per part by weight of a), and 0.005 to 0.02 part by weight of phosphorus as d) per part by weight of a).
4. A composition according to claim 2 or claim 3 wherein a) is heated concurrently with b), c) and d).
5. A composition according to any one of claims 2 to 4 wherein d) is phosphorous acid, H_3PO_3 .
6. A composition according to any one of claims 2 to 5 wherein a) is heated concurrently with b), c) and d) in proportions on an active ingredient basis of 0.15 to 0.2 part by weight of b) per part by weight of a), 0.015 to 0.02 part by weight of boron as c) per part by weight of a), and 0.01 to 0.02 part by weight of phosphorus as d) per part by weight of a).
7. A composition according to any one of the preceding claims wherein the ashless dispersant is a succinimide ashless dispersant.
8. A composition according to any one of the preceding claims wherein the ashless dispersant is a polyisobutenyl succinimide ashless dispersant formed from a polyisobutenyl succinic acylating agent and triethylene tetramine in a mole ratio of less than 2.0 but more than 1.3 moles of said acylating agent per mole of triethylene tetramine.
9. A composition according to any one of the preceding claims wherein the polyisobutenyl group of said ashless dispersant is derived from polyisobutene having a GPC number average molecular weight in the range of 700 to 1200.
10. A composition according to any one of the preceding claims wherein the ashless dispersant is a polyisobutenyl succinimide ashless dispersant formed from a polyisobutenyl succinic anhydride and triethylene tetramine in a mole ratio of 1.8

moles of said anhydride per mole of triethylene tetramine, and wherein the polyisobutenyl group of said ashless dispersant is derived from polyisobutene having a GPC number average molecular weight in the range of 850 to 1000.

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11. A composition according to any one of the preceding claims wherein c) is a boron acid.

12. A composition according to any one of the preceding claims wherein b) is an ethoxylated C₈-C₁₆ alcohol having an average of 1 to 3 ethoxy groups per molecule.

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13. A composition according to any one of the preceding claims wherein b) is an ethoxylated C₁₂ alcohol having an average of 1 to 3 ethoxy groups per molecule and wherein c) is boric acid.

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14. A lubricating oil composition which comprises from 0.1 to 99.9 parts by weight of a composition according to any one of the preceding claims and from 99.9 to 0.1 parts by weight of at least one oil of lubricating viscosity.

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15. A lubricating oil composition which comprises from 0.1 to 50 parts by weight of a composition according to any one of the preceding claims and from 50 to 0.1 parts by weight of at least one oil of lubricating viscosity.

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